Field dependence of the magnetic ordering of Cu in R_2CuO_4 (R=Nd,Sm)

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We have used neutron diffraction techniques to study the field dependence of the magnetic ordering of Cu spins in R_2CuO_4 (R=Nd,Sm) in order to distinguish between the proposed collinear and noncollinear spin structures. In the proposed collinear spin structure, there are two separate domains with the spins either along the (110) or along the ($1\bar{1}0$) directions, while in the noncollinear model there is a single domain with the alternate-layer spins along the (100) and (010) directions, respectively. If a magnetic field is applied along the ($1\bar{1}0$), strong hysteresis effects are anticipated for the collinear spin structure due to domain repopulation, while such effects are not expected for the noncollinear spin structure. Our field dependent data do not show any hysteresis effects associated with the pure Cu ordering, which strongly suggest that the noncollinear spin structure is correct for the magnetic spin configuration of the Cu spins in both compounds. Hysteresis effects in a field are observed in Sm_2CuO_4 near and below the Sm ordering temperature, and these are most likely caused by the interaction between Sm and Cu sublattices.

The magnetic properties of Cu in R_2CuO_4 (R=Nd, Pr, and Sm), which are parent materials of the electron superconductors, have been studied using neutron diffraction techniques.[1] Long range antiferromagnetic order of Cu develops in these compounds at high temperatures (~ 280 K), with a simple spin configuration in which nearest-neighbor spins within the Cu–O planes are antiparallel. The coupling between the layers, on the other hand, cancels to a first approximation, and the delicate balance of the interlayer interactions leads to rich behavior as a function of temperature and magnetic field. In particular, two spin reorientation transitions at 75 and 30 K are observed in Nd_2CuO_4, while similar reorientations are not observed in the sister compounds Pr_2CuO_4 and Sm_2CuO_4. In addition, there is a strong zero-field coupling between the rare earth ions (R^{3+}) and Cu spins in Nd_2CuO_4 while there is no indication of such a coupling in Sm_2CuO_4.

Both collinear (single-q) and noncollinear (double-q) spin structures, which are shown for Nd_2CuO_4 in Fig. 1, have been proposed for these systems.[2,3] Open and closed circles represent the $z=0$ and $z=c/2$ planes, respectively. The nearest-neighbor spins within the a–b plane have the antiparallel arrangement in both structures as shown in Fig. 1. In the collinear model, spins in adjacent planes are collinear and the magnetic symmetry is orthorhombic. There are two separate domains in this model, with the spins pointing either along the (110) or along the ($1\bar{1}0$) directions, which are shown in Figs. 1(a) and 1(b). The difference between this model and the noncollinear model is that the Cu spins in adjacent planes are rotated by $\pi/2$, and this yields a tetragonal magnetic symmetry. This noncollinear model has only one domain with the spins in the alternate layers along the (100) and then the (010) directions as shown in Fig. 1(c). This structure can be considered as the coherent addition of the two separate domains [Figs. 1(a) and 1(b)] of the collinear structure. Both polarized and unpolarized zero-field neutron diffraction data cannot distinguish between these collinear (single-q, two-domain) and noncollinear (double-q, single-domain) spin structures.[1,4–6] Similar ambiguities in spin structures have occurred in other compounds, and by applying magnetic field or stress these may be resolved.[7] In this paper, we report our field-dependent neutron diffraction measurements for Nd_2CuO_4 and Sm_2CuO_4, and these
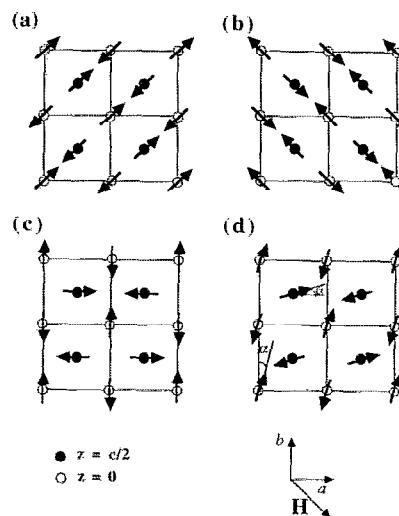

FIG. 1. Possible collinear and noncollinear spin structures for Cu spins in Nd_2CuO_4. Open and closed circles are in the $z=0$ and $z=c/2$ planes, respectively. There are two separate domains in the collinear structure. (a) Domain-1 collinear structure. (b) Domain-2 collinear structure. (c) Noncollinear spin structure. Our field-dependent data strongly indicate that the noncollinear model is correct. (d) Noncollinear spin structure in a magnetic field, which is along ($1\bar{1}0$). $\alpha=0$ at zero field [Fig. 1(c)], and α increases with increasing field. At sufficiently high fields, $\alpha \rightarrow \pi/4$ and the spin structure approaches the collinear model shown in (a).

data strongly suggest that the noncollinear spin structure is the correct one for the Cu spins.

The neutron experiments were carried out on the BT-2 and BT-9 triple axis spectrometers at the Research Reactor at the National Institute of Standards and Technology. Unpolarized diffraction data were taken using a pyrolytic graphite monochromator and filter, with an incident energy of 14.8 meV. Single crystals of Nd_2CuO_4 and Sm_2CuO_4, weighing 261 and 66 mg, respectively, were used in these experiments. The sample preparation technique can be found elsewhere.[5,8] A split-coil superconducting magnet with a vertical field capability of 7 T and a helium cryostat were employed. Crystals were mounted in the (hhl) scattering plane, with the field applied vertically, that is, in the $(1\bar{1}0)$ direction.

The two separate domains of the collinear spin structure, which are shown in Figs. 1(a) and 1(b) for Nd_2CuO_4, are equally populated at zero field. If a magnetic field is applied, spins will prefer to be perpendicular to the field because the transverse susceptibility is greater than the parallel susceptibility, and domain-1 [Fig. 1(a)] will be more favorable than domain-2 [Fig. 1(b)]. Hence, spins along $(1\bar{1}0)$ want to change their direction to (110) with increasing field (i.e., domain-1's population will increase). If the field is then removed, the two domains are energetically equivalent and thus it is not necessary for those spins to return to their original direction. Hence, strong irreversibility (hysteresis) effects are anticipated for the collinear structure and have been observed in other compounds.[9] The noncollinear spin structure, on the other hand, has only one domain, which is shown in Fig. 1(c). Since the spins will prefer to be perpendicular to the field, spins in the noncollinear structure will also rotate as shown in Fig. 1(d). The rotation angle α, which is zero at zero field, will increase with increasing field, and the spin structure at sufficiently large fields will approach the collinear structure as shown in Fig. 1(a) $(\alpha = \pi/4)$. If the field is removed, the spins will rotate back to their original direction, so irreversibility effects in the field dependence for the noncollinear structure are not expected.

Examples of the intensities of magnetic Bragg peaks for Nd_2CuO_4 ($T = 125$ K) and Sm_2CuO_4 ($T = 80$ K) as a function of field are shown in Fig. 2. Both field cooled and zero-field cooled (from 300 K) data showed similar behavior. No hysteresis effects are observed in these measurements, nor did we find any hysteresis at a series of other temperatures. The only exception to this was observed for Sm_2CuO_4 near and below the Sm ordering temperature, which we believe originates from the Sm–Cu coupling. Hence, for the Cu spins these observations strongly suggest that the noncollinear structure is the correct one in both compounds. Similar conclusions have very recently been drawn by Petigrand et al.[10] and Cherny et al.[11]

In Nd_2CuO_4, we have measured the field dependence of several magnetic Bragg peaks at a series of temperatures. With increasing magnetic field, some magnetic Bragg peak intensities decrease [e.g., $(\frac{1}{2}\frac{1}{2}1)$ at 125 K as shown in Fig. 2(a)] while other peaks increase [e.g. $(\frac{1}{2}\frac{1}{2}2)$ at 125 K]. The details will be published elsewhere.[12] Our observations

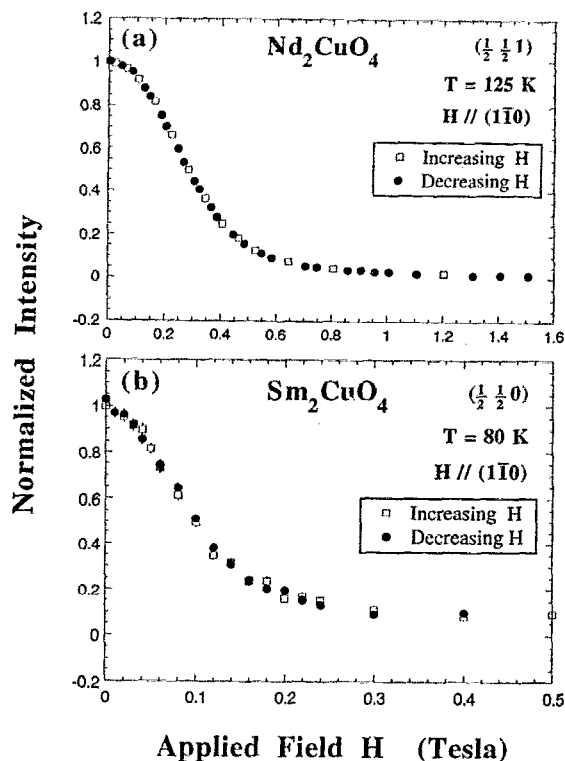FIG. 2. The magnetic peak intensities, which are normalized to zero-field intensity, as a function of magnetic field $H\|$ $(1\bar{1}0)$, for (a) Nd_2CuO_4 and (b) Sm_2CuO_4. There is no indication of hysteresis in these compounds, and this observation strongly suggests that the noncollinear spin structure is correct for Cu spins for Nd_2CuO_4 and Sm_2CuO_4.

agree very well with our conclusion that the zero-field spin structure is noncollinear. We note that two spin reorientation transitions are observed at 75 and 30 K in this system.[1-3] In the spin reorientation region (30 K $< T < 75$ K), the field-dependent behavior is reversed (i.e., with increasing field the peaks which increased in intensity for $T > 75$ K, decrease, and vice versa). These observations agree with the noncollinear structure (zero-field), which is similar to Fig. 1(c) but with spins rotated by $\pi/2$ about the c axis. Some of the field-dependent data are shown in Fig. 3.
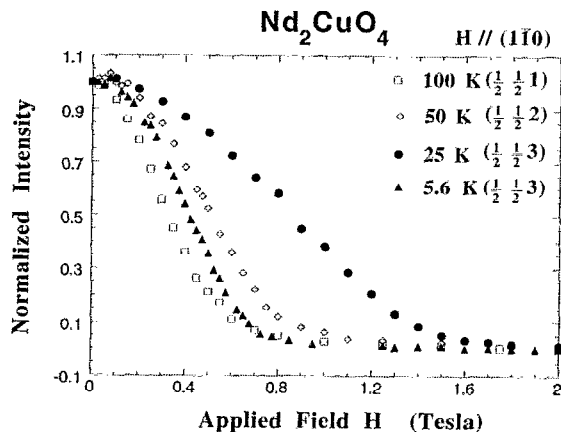

FIG. 3. The magnetic peak intensities as a function of magnetic field $H\|$ $(1\bar{1}0)$ for Nd_2CuO_4 at different temperatures.
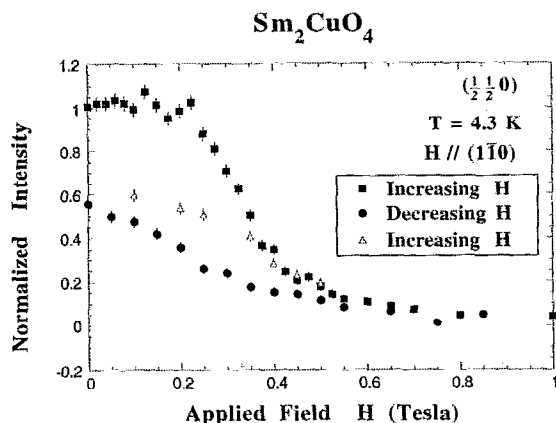
Sm₂CuO₄

FIG. 4. The magnetic peak intensity as a function of field for $H\|$ $(1\bar{1}0)$ for Sm_2CuO_4 at 4.33 K. These data show strong hysteresis. The sample was cooled in zero field from 80 K and increasing field data (solid squares) were taken first. Solid circles are for decreasing field, and the field was increased again (open triangles). This hysteresis effect, which may be due to the interaction between the Sm and Cu sublattices, is observed only near and below the Sm ordering temperature.

The field required to change the noncollinear structure to collinear increases with decreasing temperature down to 30 K, while below 30 K it decreases. These data are in good agreement with previous magnetization[11] and neutron scattering measurements.[10] There is a strong interaction between the Nd and Cu spins below 30 K,[1,6] and the ordered Cu sublattice induces the Nd spins to align. The Nd spins may be rotated to a collinear structure with a smaller field, and the Cu spins may also rotate due to the interaction with the Nd spins. This may be the reason that the field required to change the spin structure at 5.6 K is smaller than what is required at 25 K.

For the Sm_2CuO_4 system the Cu spins order antiferromagnetically ($T_N = 280$ K[5]) and the field-dependent data also show no hysteresis at high temperatures ($T \gtrsim 20$ K) as shown in Fig. 2(b). We note that the field dependence of Sm_2CuO_4 is similar to that measured for Nd_2CuO_4 in the spin reorientation region, as expected since the spin structures are identical in these two regimes. This structure is similar to the one shown in Fig. 1(c) but with spins rotated by $\pi/2$ about the c axis.

A strong hysteresis effect has been observed at low temperatures in Sm_2CuO_4, and some data are shown in Fig. 4. Measurements were taken after cooling the sample to 4.3 K in zero field. The intensity of the $(\frac{1}{2}\frac{1}{2}0)$ magnetic

peak drops by about 40% once the field is increased to 7 T and returned to zero, while the $(\frac{1}{2}\frac{1}{2}1)$ magnetic peak intensity increases. The virgin intensity can be recovered by warming the sample above 10 K and then cooling in zero field again. Such measurements were repeated at $T = 8$, 10, 12, and 20 K after warming the sample to 40 K and returning it back to T. If the field is increased to 7 T and then returned to zero the intensity of the $(\frac{1}{2}\frac{1}{2}0)$ peak drops only by $10\pm4\%$ at 8 K, and this hysteresis effect completely goes away at 20 K. We note that the Sm spins order at 5.95 K with a completely different spin structure than the Cu, and the ordered Sm moment is along c axis.[13] Thus this hysteresis effect might be due to the interaction between the Sm and Cu sublattices, even though there is no indication of such an interaction at zero field.[13] We remark that for Nd_2CuO_4 the Nd[11] and Cu spins have the same noncollinear structure and moment directions. Hence the Sm and Cu sublattices' influences on each other are different, and they may be affected by magnetic field, causing the hysteresis effect.

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